

# ABSOLUTE SPIN MAGNETIC MOMENT OF $\text{FeF}_2$ FROM HIGH-ENERGY PHOTON DIFFRACTION

Separating spin and orbital contributions to the magnetic moment in a magnetically ordered system has long been a challenge. For the first time, spin and orbital contributions were separated using high-energy x-ray diffraction in combination with results obtained by other methods, e.g., neutron diffraction and antiferromagnetic resonance. The investigated material was  $\text{FeF}_2$ , a system that shows a significant orbital momentum. The reliability of this method shows clearly that such a separation is possible to a precision better than 2%.

The separation of the spin and orbital contribution to the magnetic moment in magnetic materials has been a challenge for a long time in experimental condensed-matter physics. The separate knowledge of both components provides important information about spin-orbit and Coulomb interactions and crystal field effects in the solid. Several methods can be used to probe the total magnetic moment. But a separation of the components is rather complicated and a real challenge. In the last few years new methods using x-rays have been developed that offer great opportunities in this direction.

We discuss results of a magnetic diffraction experiment on antiferromagnetic  $\text{FeF}_2$  with 115 keV photons [1]. At this energy, the magnetic scattering cross section only depends on the spin component perpendicular to the diffraction plane and is independent from the polarization of the x-rays. Combined with antiferromagnetic resonance and neutron diffraction results, this offers the possibility to separately determine the spin and orbital contribution to the magnetic moment.

For x-rays, the magnetic signal is 6 to 8 orders of magnitude smaller than the signal from conventional charge scattering. This is mainly the case because magnetic x-ray scattering is a relativistic process. Therefore, a magnetic signal can be measured only at positions where no additional charge

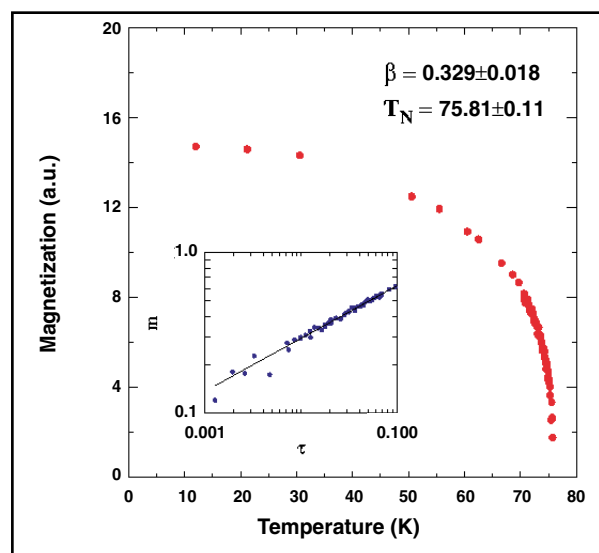


FIG. 1. Sublattice magnetization from the magnetic (300)-reflection of  $\text{FeF}_2$  as a function of temperature. The inset shows the reduced sublattice magnetization as a function of the reduced temperature in double logarithmic scale. The line is a fit of a power law to the data.

scattering occurs. This is possible only for antiferromagnets, where a doubling of the magnetic unit cell occurs with respect to the chemical unit cell, or the magnetic reflections appear at positions where the charge signal is extinct, as is the case for  $\text{FeF}_2$ . The simple cross section, which is proportional to the spin only, is valid for energies larger than 80 keV. This has been demonstrated experimentally by dif-

fraction experiments from  $\text{MnF}_2$  [2]. For neutrons, the diffracted intensity is proportional to the linear combination  $L(Q) + 2S(Q)$  of both the Fourier transforms of the spin  $[S(Q)]$  and orbital angular  $[L(Q)]$  momentum. Thus, by combining the results of high-energy x-ray and neutron diffraction experiments, orbital and spin contributions can be separated without further theoretical assumptions.

The experiment was conducted at the wiggler beamline of BESSRC-CAT at the Advanced Photon Source at Argonne National Laboratory [3]. The crystal was mounted in a closed-cycle cryostat and allowed to reach a minimum temperature of 12 K.

The temperature dependence of the magnetic intensity was measured at the purely magnetic (300)-reflection (see Fig. 1). The magnetic moment is increasing with decreasing temperature, i.e., measured intensities become stronger. In addition, the critical region just below the phase transition was investigated. Here, the nature of the phase transition can be studied. A power law for the reduced sublattice magnetization as a function of reduced temperature,  $\mu = \tau^\beta$ , was fit to the data in the temperature range between 70 K and 75 K, resulting in a critical exponent of  $\beta = 0.329(18)$  (inset of Fig. 1). This value agrees very well with the  $\beta = 0.325(5)$  obtained from NMR measurements [4] and Mössbauer spectroscopy [5]. It is in perfect agreement with the value  $\beta = 0.326$  calculated for the 3-d Ising model, where the alignment of the magnetic moment is limited to one direction, e.g., the c-axis.

For the determination of the spin magnetic moment, magnetic intensities have been measured at the positions (100), (300), and (500). Although, in the case of  $\text{FeF}_2$ , the magnetic signal is 6 orders of magnitude smaller than the conventional charge scattered signal, peak intensities of up to 6000 cps in the reflection could be obtained, because of the very good crystal quality. The magnetic intensities have been normalized to the intensities of the (200) and (400) charge reflections, where the structure factors are known very accurately from  $\gamma$ -ray experiments [6]. The absolute magnetic structure factors, which are obtained after the necessary corrections detailed in Ref. 1, are shown in Fig. 2. An absolute

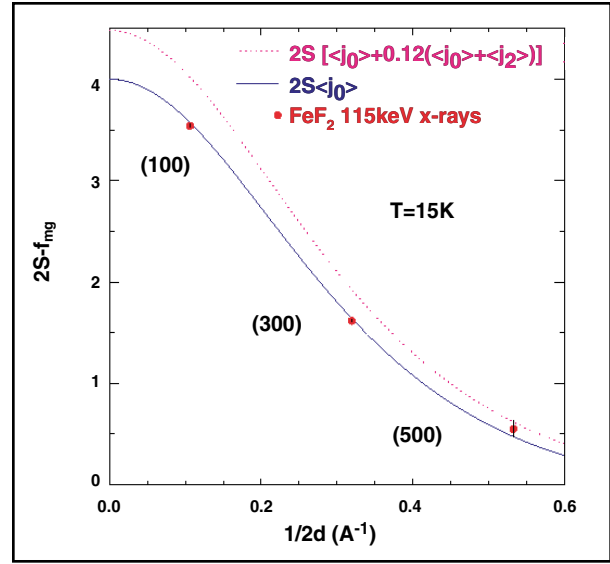


FIG. 2. Absolute magnetic structure factor as a function of  $\sin \theta/\lambda$ . The solid points show the experimental data from the magnetic (100), (300), and (500) reflections. The solid line shows the pure spin form factor. The dashed line shows the expected behavior from neutron diffraction.

spin magnetic moment of  $\mu = 4.01(5)\mu_B$  could be determined by extrapolation to  $Q = 0$ .

This result supports the assumption that in  $\text{FeF}_2$  the spin is  $S = 2$  as in the free ion  $\text{Fe}^{2+}$ . The 12% higher magnetic moment of  $\mu = 4.46\mu_B$ , measured in a polarized neutron diffraction experiment [7], therefore results from an additional orbital component.

For the first time, a spin-orbit separation was performed with high-energy x-rays in combination with results obtained by other methods, e.g., neutron diffraction and antiferromagnetic resonance, on a system that shows a significant orbital momentum. In other studies done previously with this method, the system either has not shown an orbital contribution or a separation was problematic because of the nonuniform distribution of the magnetic domains in the crystal. Since the form factor as a function of  $Q$  for the pure spin, on the one hand, and for spin and orbital contribution on the other, are very similar in form, the obtained information about the absolute magnetic form factor is essential for a precise separation. Our result underlines the reliability of this method and shows clearly that such a separation is possible to a precision better than 2%.

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